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Monitoring volatilization products using Residual Gas Analyzers during MeV ion beam irradiations

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ABSTRACT

The use of Residual Gas Analyzers (RGAs) during irradiation experiments can provide valuable information when incorporated into experimental end-stations. The instruments can track the volatilization products of beam-sensitive materials, which may ultimately aid researchers in selecting appropriate flux values for conducting experiments. Furthermore, the type of gaseous species released during an irradiation can be monitored directly, which may lead to new insights into the radiolysis and/or heating mechanisms responsible for gas evolution. A survey of several classes of materials exposed to extremes in particle flux is presented to show how RGA instrumentation can be incorporated to qualitatively assess ion-solid interactions in a variety of fields.

1. Introduction

Common to many modern vacuum and accelerator systems are Residual Gas Analyzers (RGAs). RGAs are mass spectrometers used for monitoring vacuum conditions and are especially useful for identifying leaks. For instance, in a typical vacuum system operating in the 10^{-8} Torr range, an RGA readily measures the $\sim 10^{-9}$ partial pressures of air, water, and oils, and is readily configured into a leak detection mode for helium measurement. However, if an RGA is appropriately designed into experimental end-stations, it can become a useful instrument for measuring gaseous species that may become volatilized due to ionization and beam heating during irradiation experiments. Two possible applications for RGAs in irradiation experiments include determining the proper beam flux for conducting accelerated irradiation experiments, and direct measurement of radiolysis (ionization) effects in materials.

Accelerated irradiation studies are experimental irradiations performed at dose rates in excess of the dose rates associated with the physical phenomena they are meant to simulate: such as in nuclear reactors, radioactive waste storage, and space weathering [2,14,21]. Whereas the energy and fluence in irradiation experiments are typically predetermined, the flux is often not. Many experimentalists overlook this crucial variable, as the flux, or beam current, is oftentimes determined by what the equipment is capable of outputting on any given day. Researchers benefit from stable, high-current irradiations as they can save valuable time over the course of a long irradiation campaign, particularly when considering experiments lasting days and weeks. Furthermore, it is easy to reconcile the economics of an irradiation that could be performed over the course of eighty hours, or *tweaking* a couple of knobs in order to complete the experiment in one long working day.

Most ion irradiation experiments typically use keV or MeV beams with nanoamp to microamp beam currents, resulting in fractions of a watt-like energy deposition. Even these seemingly innocuous wattages can elevate the temperature in the irradiated region, as the volume of energy deposition is small; ion ranges are typically on the order of nanometers to tens of microns in depth for most materials. Limiting the temperature gain in the target is critical, as elevated temperatures may result in deleterious effects such as volatilization and undesired defect recombination or dynamic annealing [7,11,17]. This limits the flux of particles which can be used without considering extensive mechanisms for heat removal such as liquid interfaces and other cooling schemes during irradiation [16].

In addition to tracking the role of beam flux during an irradiation, the RGA can be used to understand radiation degradation and radiolysis in materials. For instance, the technique has been used to simulate α-particle exposure in polymers that come into close contact with actinides [6,18,22]. A novel analogous example is understanding the effects of high-energy protons from solar flares on precursor planetary material. Stars such as our Sun were far more active proton emitters during their early evolution. It is possible that chemical alteration of primitive solar material could be simulated in experiments similar to those above,

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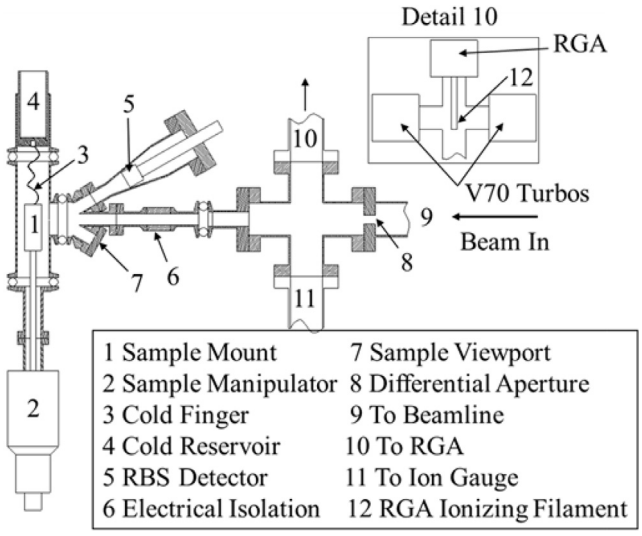

Fig. 1. An experimental end-station capable of performing both traditional ion beam analysis and RGA studies. Volatile products generated on the target located at (1) are pulled in an opposite direction to the beam towards the turbo pumps for the RGA and beamline; the aperture at (8) can be used to restrict loss to the beamline. To assure all gasses generated from exposure were measured by the RGA, a beam transparent window could be inserted at (8) to isolate the chamber completely from the remainder of the accelerator, increasing the sensitivity on the spectrometer.

with chemicals released from the target material being tracked with the RGA.

In this report, we present several examples of ion irradiation experiments wherein we used an RGA to monitor radiolysis effects and target volatilization. Our examples involve multiple classes of materials.

2. Experimental

A series of ion beam irradiations were performed using 2 MeV H^+ ions (University of Wisconsin-UW) and 5.5 MeV He^{++} ions (Los Alamos National Laboratory-LANL). Fig. 1 shows a typical experimental setup. A Stanford Research Systems (SRS) 300 amu (atomic mass unit) RGA was connected in series with the end-station and the accelerator beam line for the LANL experiments. Twin 70 *l/s* turbo pumps are adjacent to the ionizing filament as can be observed in detail 10 of the figure. For the experiments conducted at UW, the RGA was mounted on the side of the end-station using a single 70 *l/s* pump adjacent to the filament. In both configurations, some portion of the volatilized products are pulled past the RGA filament due to the local pressure gradient created by the turbo pumps. The SRS RGA has manufacturer detection limits of 5×10^{-11} Torr using the instrument's Faraday cup, and 5×10^{-14} when the optional electron multiplier is used. The maximum operating pressures for the Faraday cup and electron multiplier are 10^{-4} and 10^{-6} Torr respectively. The instrument is capable of monitoring up to 10 different amu species simultaneously, with the data being output in multiple pressure-versus-time configurations. The data presented in this report typically entails only one or two of the volatilized products for a given sample, and is displayed with the partial pressure (Torr) on the ordinate, with time on the abscissa. It should be noted that the mass filter selects species based on their mass over charge ratio (m/q), and that data displayed represents a major element in the target. The targets were mechanically mounted on an electrically isolated, un-biased stainless steel stage, with charge collected using a Brookhaven Instruments Corporation (BIC) current integrator. The current measured on the isolated stage agreed well with a pre-chamber Faraday cup.

Materials examined include: (1) a polished slab of Portland cement and (2) a zone refined nickel slab, each irradiated with un-scanned 2 MeV H^+ ions to simulate neutron exposure in a reactor environment; (3) a Kapton® (polyimide) film irradiated with scanned 5.5 MeV He^{++} ions to simulate α-particle exposure from an actinide; and (4) a sulfide mineral irradiated with un-scanned 2 MeV H^+ ions to simulate meteorite precursor material exposed to solar flare particles during stellar evolution. Post irradiation analysis techniques employed in this study include scanning electron microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDXS) on the sulfide mineral using a Phenom Pro-X SEM, and Thermal Gravimetric Analysis (TGA) on the cement using a TA Instruments Q500. Experimental irradiation conditions are summarized in Table 1.

3. Results and discussion

3.1. Cement example

Fig. 2 shows an RGA mass scan for microcrystalline cement irradiated with 2 MeV protons using two different ion fluxes. The initial background pressure of water for the cement is over two orders of magnitude higher than the other materials used in this study. Fig. 2 shows that a flux of 4.7×10^{12} H^+/cm^2-s was applied at an elapsed time of 15 s and stopped after a minute of irradiation. A second irradiation was performed using a flux of 9.4×10^{12} H^+/cm^2-s and started at an elapsed time of 120 s, and stopped 140 s later. The irradiation was performed over a 0.04 cm^2 area with respective beam currents of 30 and 60 nA.

A considerable difference in water loss is evident between the two flux values. Understanding and limiting water loss should be critical in performing accelerated aging studies in cement. Both radiolysis and heating effects during irradiation may result in water loss and subsequent mechanical property degradation of concrete structures [5,12].

Gas generation in the case of cement via radiolysis is accomplished by the dissociation of water and the subsequent formation of liberated molecular species. Limited literature exists which describes the radiolysis of cement. However, a specific concern in these scenarios is the buildup and explosion hazard of H_2 gas generated from radioactive materials sealed in cement casks. Siskind [20] determined that H_2 and H_2O are generated by the radiolysis of trapped pore water in the cement

Table 1
Target details and experimental irradiation conditions.

Material	Target Geometry	Ion & Energy	Facility	Irradiation Area	Beam Current	Flux/Fluence
Portland Cement	Polished Slab 48 mm^2 × 4 mm thick	2 MeV H^+	UW	0.04 cm^2	30 nA 60 nA	4.7×10^{12} H^+/cm^2 9.4×10^{12} H^+/cm^2
Nickel	Zone Refined Boule 150 mm^2 × 0.7 mm thick	2 MeV H^+	UW	0.1 cm^2	1 μA 6 μA 8 μA 10 μA	6.2×10^{13} H^+/cm^2 3.7×10^{14} H^+/cm^2 5.0×10^{14} H^+/cm^2 6.2×10^{14} H^+/cm^2
Kapton© HN	Film 100 mm^2 × 0.13 mm thick	5.5 MeV He^{++}	LANL	1 cm^2 (0.09 cm^2 Beam Spot)	25 nA	2.7×10^{13} He^{++}/cm^2
FeS_2	Irregular Mineral Slab ~64 mm^2 × 2 mm thick	2 MeV H^+	UW	0.1 cm^2	1 μA 1.5 μA	6.2×10^{13} H^+/cm^2-s 9.5×10^{13} H^+/cm^2-s